Appl. No. 10/537,973

Amendment dated June 9, 2008

Applicants' Response to the Office Action mailed December 7, 2007

REMARKS/ARGUMENT

Amendments to the Claims

Claim 13 has been amended and limited in scope by incorporation of some of the heterogeneous catalysts claimed in previously-presented Claim 24, with the remainder of catalysts from Claim 24 being included in new Claim 26, as suggested by the Examiner, thus resulting in the cancellation of Claim 24. In none of these cases is any new matter added, nor any material deleted from the Claims as a whole, with all such changes fully supported by the original Claims and Specification. Entry of these amendments and continued examination based on the same is, therefore, respectfully requested.

The instant invention is directed to a countercurrent, liquid-phase, reduced-heat process for the continuous esterification of C1-C22-fatty acids, including mono- and polybasic carboxylic acids, such as formic acid, acetic acid, adipic acid, dodecanedioic acid, citric acid and isophthalic acid, aliphatic fatty acids, such as caproic acid, caprylic acid, 2-ethylhexanoic acid, capric acid, lauric acid, isotridecanoic acid, myristic acid, palmitic acid, palmitoleic acid, stearic acid, isostearic acid, oleic acid, elaidic acid, petroselic acid, linoleic acid, linolenic acid, elaeostearic acid, arachic acid, gadoleic acid, behenic acid, and erucic acid, and technical mixtures thereof. including, e.g., coconut oil fatty acid, palm oil fatty acid, palm kernel oil fatty acid, and tallow fatty acid, and those obtained in the working up of fatty acid mixtures of other natural origins, with one or more linear or branched C1-C10-monoalkanols, preferably C1-C8-monoalkanols, more preferably isopropanol or 2-ethylhexanol, C2-C5-dialkanols or C2-C5-trialkanols, such as glycerol, ethanediol, propane-1,2-diol, propane-1,3-diol, butanediol, pentanediol, and isomers and semi-esters thereof, preferably C2- or C3-dialkanols or -trialkanols, more preferably glycerol, or mixtures of two or more of the monoalkanols, dialkanols and/or trialkanols, in the presence of one or more heterogeneous catalysts, selected from organic or inorganic, acidic or basic ion exchangers, acidic clays, and zeolites, preferably including specially-worked-up bleaching earths and catalysts based on transition metals, more preferably acidic cation exchangers. The initial/partial esterification occurs in a preliminary (preferably fixed-bed) reactor at 50-to-150.

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preferably 80-to-120°C, and 1-to-10, preferably 1-to-5 bar pressure, containing the catalyst(s), which is followed by a separation unit, such as a falling-film evaporator, phase separator, or membrane unit, for reducing the viscosity of the reaction mixture and removing the water of reaction in order to displace (toward greater ester formation) the reaction equilibrium. Further esterification then occurs in a reaction column, charged with the heterogeneous catalyst(s) directly on the column (e.g., sieve or bubble) plates, operated at 50-to-200, preferably 80-to-150°C, and 0.1-to-10, preferably 0.1-to-5 bar pressure, preferably into which column nitrogen is fed from the lowermost plate, in order to increase the vapor load in the lower part of the column, remove additional water of reaction, deodorize the reaction mixture, and create a more favorable acid-to-alcohol ratio, and surmounted on which column is a rectifying section.

Claims 13-23 have been rejected under 35 USC 112, first paragraph, as the Specification is alleged to "not reasonably provide enablement for any catalysts" other than AmberlystTM 17 macroreticular strong acid cation exchange resin.

Applicants' amendment to Claim 13, limiting the scope of "heterogeneous catalyst" to some of those included in previous Claim 24 is believed to satisfy and overcome the Examiner's concern related to the use of the term.

While Applicants have happened to have employed Amberlyst^M17 macroreticular/macroporous strong acid cation exchange resin as the heterogeneous catalyst in their two Examples and mentioned it as an example of an acidic cation exchanger in a particular embodiment of their invention on page 9, lines 24-26 of the Specification, this catalyst is, in no way, essential to their invention or an aspect on which it depends. The instant invention certainly involves a heterogeneous catalyst, and AmberlystTM17 strong acid cation exchange resin is one of our preferred examples, but, like temperatures, pressures, amounts of catalyst, initial and operating costs of the process, the particular reactants and the end product desired, not all of these variables

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have to be included in extensive Examples, each with a variety of these variables, in order to demonstrate viability of Applicants' novel process to the reasonably-skilled practitioner.

The use of a heterogeneous catalyst in an esterification process is well-established (see, e.g., United States Patent 6, 245,727 B1 (Gutsche et al) and §14.10.4.2.1, p.3329, of 14.10.4 "Heterogeneous Catalysis in Oleochemistry" from the 8-volume Handbook of Heterogeneous Catalysis, Second Edition, G. Ertl et al, eds., Wiley-VCH Verlag GmbH & Co. KGaA [enclosed herewith]), as is the use, in esterification reactions, of catalysts with different properties that may be provided or mimicked by heterogeneous catalysts, according to "Chemiker-Ztg./Chem. Apparatus 87", No. 18, by H. Stage (1963), pp. 661-666, and organic chemistry textbooks, such as Organic Chemistry. Sixth Edition, by Morrison and Boyd, Prentice Hall (1992) e.g., §§6.11, 19.9, 19.16 and 20.15 and Organic Chemistry, by F.A. Carey, The McGraw-Hill Companies, Inc. (1996) e.g., §§15.8, 15.17, 20.7 and 27.5).

As "the level of skill in...esterification is high", as pointed out by the Examiner on page 9 of the instant Office Action, selection of the appropriate heterogeneous catalyst for an esterification reaction with particular reactants and circumstances would be well within the competence of the reasonably-skilled practitioner-whether a B.S.-degree chemist or a Ph.D. chemist or chemical engineer. Such an artisan might employ some limited trial and error in order to optimize the selection of the best heterogeneous catalyst, but he/she would not have to engage in extended or undue experimentation in order to make such a selection. The selection of an esterification catalyst for particular reactants and operating conditions in a very widely-practiced art is simply not as mysterious as suggested, and the artisan would certainly expect that many different heterogeneous catalysts, selected according to applicability for the particular reactants and circumstances, would work successfully in Applicants' process.

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With Applicants' amendment to Claim 13 and in view of this discussion, Applicants respectfully believe they have fully satisfied and overcome this rejection, and, therefore, respectfully request that it be reconsidered and withdrawn.

Claim 24 has been rejected under 35 USC 112, second paragraph, as indefinite for "not clearly set[ting] forth the metes and bounds of the patent protection desired" as containing a narrowing of useful esterification catalysts from an already-rejected broad range.

Applicants believe that they have adopted the Examiner's suggestion, and by adding a new Claim 26, have satisfied and overcome this rejection.

Reconsideration and withdrawal of the rejection is, therefore, respectfully requested.

Claims 13-25 have been rejected under 35 USC 103(a) as unpatentable over Kahsnitz et al (US 5,177,229) in view of Bremus et al (US 5,008,046).

United States Patent 5,177,229 (Kahsnitz et al), claiming common priority with EP 0 474 996 A1 discussed on page 2, lines 21-28 of the present Specification, describes an esterification process comprising reacting an alcohol, preferably a C₁-C₆-alcohol, such as methanol, ethanol, propanol, isopropanol, butanol, pentanol, hexanol, and cyclohexanol, preferably methanol or ethanol, and an acid, preferably a C₆-C₂₀-carboxylic acid, such as hexanoic, decanoic, dodecanoic, tetradecanoic, palmitic, linoleic, stearic, benzoic, adipic, dodecanedioic, citric, or isophthalic acid, particularly C₁₀-C₁₆-aliphatic monocarboxylic acids, preferably at 0.2-to-6, more preferably at 1-to-5 equivalents of alcohol per equivalent of acid, by liquid-phase equilibrium reactions on ion exchangers, preferably commercially-available acidic cation exchange resins, in a prereactor at up to 120, preferably at 40-to-100°C, followed by a rectifying column of 5-to-30 theoretical plates, from which, preferably, the entire liquid phase on a column plate is withdrawn from the side, and directed to one of from 2-to-5 external reactors, to which a portion of the alcohol or the

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acid, preferably the lower-cost alcohol, in 0.2-to-5, preferably 1-to-3 equivalents per equivalent of acid, is also fed directly. From 40-to-85 mole % of the mixture leaving the external reactors is evaporated, with the vapor phase being added back to the column at the first plate above the earlier discharge point, with the remaining liquid phase being returned to the column to the next lower plate from the discharge point.

While disclosing an ion exchange resin-catalyzed, liquid-phase esterification process, the reaction described in the Kahsnitz et al patent simply is not conducted in a countercurrent flow and is not continuous. Thus, it neither teaches nor suggests Applicants' process.

United States Patent 5,008,046 (Bremus et al), cited on the Kahsnitz et al patent, describes a continuous, liquid-phase esterification, intended to reduce the dehydration of the alkanols, of C2-C24-fatty acids, or mixtures of such acids of natural (e.g., vegetable, animal and/or seaanimal) origin or of synthetic origin with C1-C5-monoalkanols, preferably ethanol, propanol, butanol, pentanol and isomers thereof, preferably in a molar ratio of less than 2:1, preferably from 1.1:1-to-1.8:1, more preferably 1.2:1-to-1.4:1 monoalkanols-to-fatty acids, or C2- or C3dihydric alcohols, such as ethylene glycol or propylene glycol in molar ratios adapted accordingly, in the presence of catalysts, preferably selected from sulfuric acid, toluenesulfonic acid, chlorosulfonic acid, and methylsulfonic acid, more preferably p-toluenesulfonic acid, resulting in elimination or substantial reduction of dehydration of the alkanols when branched monoalkanols are used. The reactants are circulated in countercurrent contact, for at least 20 minutes, in a single multi-plate reaction (e.g., sieve- or, preferably, bubble-plate) column at 190. preferably at from 120-to-145°C, at a head pressure of 200-to-900, preferably 700-to-900 hPa, with the catalysts and heated acids introduced at the top of the column and the alkanols introduced in the vapor phase below the lowest plate in the column, with excess water in the water/alkanol mixture removed from the rectifying section on the top of the column being eliminated to prevent the entrainment of fatty ester in the head product and the remaining

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water/alkanol mixture, optionally after separation of excess water, at least partly returned as an azetrope to the reaction column.

The Bremus et al Patent does disclose a countercurrent esterification process, but again, does so in one step, and, in addition, uses homogeneous catalysts. Thus, the reference neither teaches nor fairly suggests Applicants' invention.

Clearly, the Bremus et al patent neither discloses nor fairly suggests Applicants' process, and further does not suggest deficiencies that would lead a chemist to somehow combine the Kahsnitz et al patent with the Bremus et al patent in order to cure those deficiencies. With hindsight, selected elements from each patent could be combined to approach several elements of Applicants' novel process, but no combination would result in a continuous esterification process. Clearly, therefore, neither reference alone, nor any possible combination of the references either teaches or fairly suggests Applicants' novel and unobvious process.

Reconsideration and withdrawal of this rejection is, therefore, respectfully solicited.

Believing that the Application is now in condition for allowance, Applicants earnestly solicit such favorable action of the Examiner, and respectfully request that a timely Notice of Allowance be issued in the prosecution of this Application. Appl. No. 10/537,973

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If any further questions do remain which may be resolved by a telephone interview, however, the Examiner is respectfully requested to telephone Applicants' undersigned Attorney.

Respectfully submitted,

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Enclosure: §14.10.4, "Heterogeneous Catalysis in Oleochemistry" from the 8-volume <u>Handbook of Heterogeneous Catalysis</u>, Second Edition, G. Ertl et al, eds., Wiley-VCH Verlag GmbH & Co. KGaA